# Biopolymeric Sustainable Materials and their Emerging Applications

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## Abstract

Advancements in polymer science and engineering have helped the scientific community to shift its attention towards the use of environmentally benign materials for reducing the environmental impact of conventional synthetic plastics. Biopolymers are environmentally benign, chemically versatile, sustainable, biocompatible, biodegradable, inherently functional, and ecofriendly materials that exhibit tremendous potential for a wide range of applications including food, electronics, agriculture, textile, biomedical, and cosmetics. This review also inspires the researchers toward more consumption of biopolymer-based composite materials as an alternative to synthetic composite materials. Herein, an overview of the latest knowledge of different natural- and synthetic-based biodegradable polymers and their fiber-reinforced composites is presented. The review discusses different degradation mechanisms of biopolymer-based composites as well as their sustainability aspects. This review also elucidates current challenges, future opportunities, and emerging applications of biopolymeric sustainable composites in numerous engineering fields. Finally, this review proposes biopolymeric sustainable materials as a propitious solution to the contemporary environmental crisis.

**Keywords:** Biopolymers, Biodegradable polymers, Biodegradability, Biocomposites, Biomedical applications

## List of abbreviations

| | |
|---|---|
| ACC | All-cellulose composite |
| AEMO | Acrylate epoxidized mustard oil |
| APHAS | American Public Health Association standard |
| ASTM | American society for testing of materials |
| BF | Bamboo fiber |
| BG | Bioglass |
| BOD | Biochemical oxygen demand |
| CA | Cellulose acetate |
| CB | Composite bioplastic |
| CCG | Chemically converted graphene |
| CG | Cashew gum |
| CMC | Carboxymethyl cellulose |
| CNC | Cellulose nanocrystals |
| CNT | Carbon nanotube |
| COVID-19 | Coronavirus disease 2019 |
| DMT | Dimethyl terephthalate |
| DS | Degree of substitution |
| ES | Extruded sheets |
| FCC | Fiber cement composites |
| FFF | Fused filament fabrication |
| GCF | Green Coconut Fiber |
| GelMA | Methacrylated gelatin |

| | |
|---|---|
| HA | Hyaluronic acid |
| HAP | Hydroxyapatite |
| KF | Kenaf Fiber |
| LCA | Life cycle assessment |
| MB | Mater-Bi® |
| MC | Microcrystalline |
| MOSF | Moringa Oleifera (Sahajana) seed filler |
| MSCs | Mesenchymal stem cells |
| MNP | Melanin nanoparticles |
| MWCNT | Multi-walled carbon nanotube |
| NFs | Natural fibers |
| nHAP | Nanohydroxyapatite |
| NPs | Nanoparticles |
| NR | Natural rubber |
| NVP | N-vinyl-2-pyrrolidone |
| OPEFB | Oil palm empty fruit bunch |
| PA | Polyamide |
| PBAT | Polybutylene adipate terephthalate |
| PBH | Polyhydroxy butyrate |
| PBSA | Poly(butylene succinate-co-butylene adipate) |
| PCL | Polycaprolactone |
| PDO | Propanediol |
| PE | Polyethylene |
| PEO | Poly (ethylene oxide) |
| PET | Polyethylene terephthalate |
| PGA | Poly(glycolic acid) |
| PHA | Poly(hydroxyalkanoate) |
| PHB | Poly(hydroxybutyrate) |
| PHBV | Polyhydroxybutyrate-valerate |
| PBS | Poly(butylene succinate) |
| PLA | Polylactic acid |
| PLLA | Poly-l-lactic acid |
| PLGA | Polylactide-co-glycolide |
| PP | Polypropylene |
| PTA | Pure terephthalic acid |
| PTT | Polytrimethylene terephthalate |
| PVA | Polyvinyl alcohol |
| PVAc | Polyvinyl acetate |
| PWBF | Plain woven banana fabric |
| rPP | Recycled polypropylene |
| RS-g-PMMA | Rice straw-g-poly methyl methacrylate |
| SB | Simple bioplastic |
| SEM | Scanning electron microscope |
| SPNCC | Sugar palm nanocrystalline cellulose |
| SPS | Sugar palm starch |
| TPS | Thermoplastic starch |
| UV | Ultraviolet |
| WPC | Wood Plastic composite |

## 1. Introduction

Nowadays, the extensive use of conventional polymers, including polyethylene (PE), thermoset epoxy resins, and polyethylene terephthalate (PET) are polluting our environment [1]–[3]. More importantly, their role in the life of both flora and fauna have some serious negative impacts due to their stability against microorganisms, humidity, and temperature. Thus, pilling up a huge amount of plastic waste [4]–[6]. Additionally, there are rising global

health concerns among the scientific community about toxic chemicals, including bisphenol A, phthalates, or polychlorinated biphenyls, which are employed for the production of plastics [7]. Thus, plastic waste management is a major environmental issue and requires suitable methods and materials for plastic decomposition [8]. Other synthetic polymers are extensively consumed globally, and it is estimated that the non-biodegradable polymer waste will reach a volume of 25 billion metric tons by 2050. Additionally, the production of plastics has exceeded 8 billion tons of plastic in the last seventy years [9]–[11]. From 2020 to 2023, the growth of natural and synthetic plastics will be 13 % per annum as per estimation. Thus, synthetic polymers pose a risk to the environment due to their exceptionally stable nature, which causes the accumulation of waste [12]. To address the issue of plastic waste, the recycling of synthetic plastics is a sustainable environmental solution. However, the recyclable polymers possess lower mechanical characteristics and cannot be employed for high-end applications [13]–[15]. Landfilling of these materials should be given the least priority as prescribed by European Union (EU) waste management directives. Therefore, recently, more emphasis was given to the use of naturally available polymers that overcome plastic waste issues due to their biodegradable nature [16]–[18].

The utilization of biodegradable polymers or biopolymers is a prominent alternative to overcome the consumption of synthetic plastic [19]. These bio-based polymers, inevitably key biopolymers derived from renewable sources including biomass, corn, sugarcane, and molasses, have gained significant attraction. Upon degradation process, these polymers reduce their pH and molecular weight as well as increase their crystallinity [20]. These polymers are renewable materials which undergo rapid degradation through natural micro-organisms without releasing any toxic or distinguishable residue under appropriate conditions, including moisture, oxygen, and temperature [21]. The reinforcement of different natural fillers/additives significantly enhances the physical as well as mechanical properties of these biomaterials [22]. These polymers are becoming indispensable and applications of biopolymers have been greatly enhanced in the last decade [23]–[25]. These biopolymers are vastly applied in broad spectrum of engineering applications such as pharmaceutical, food packaging, electronics, textile, agriculture, construction, biomedical, aerospace, and automotive industries [26]–[28]. Bio-based polymeric materials are classified into three diverse groups on the basis of their sources and production method, as presented in **Table 1**. The first group incorporates natural polymers, including polysaccharides, proteins, and lipids extracted from animals or plants. In the second group, synthetic polymers are developed through different ring-opening polymerization or condensation methods by using bio-based monomers [29]–[31]. These polymers include polycaprolactone (PCL), polybutylene adipate terephthalate (PBAT), polylactic acid (PLA), poly(butylene succinate-co-butylene adipate) (PBSA), and poly(butylene succinate) (PBS) [32]–[34]. Whereas the third group contains polymers including poly(hydroxybutyrate) (PHB), poly(hydroxyalkanoate) (PHA), and polyhydroxybutyrate-valerate (PHBV), which are developed through genetically modified bacteria or micro-organisms [35]–[37].

**Table 1** Classification of biopolymeric materials

| Classification | Source | Biopolymers | Ref. |
|---|---|---|---|
| Polypeptides/Proteins | Animals | Fish protein, collagen, silk, gelatin, casein, whey | [38] |
| | Plants | Soy protein, zein, wheat gluten | [39] |
| Polysaccharides | Animals | Hyaluronic acid, chitosan/chitin | [40] |

| | | | |
|---|---|---|---|
| | Plants | Starch, agar, cellulose, carrageenan, pectin, alginate | [41] |
| | Lipids | Fatty acid, carnauba wax, beeswax, oil | [42] |
| Biopolymers produced through microorganisms | Bio-derived monomers | PLA, PGA | [43] |
| | Microbial polyesters | PHA, PCL, PHB, and PHBV | [44] |
| | Bacterial | Cellulose, gelian, dextran, Xanthan, polygalactosamine | [45] |
| Synthetic | - | PVA, PVAc | [46] |
| | Aliphatic polyesters | PLA, PGA, PCL | [47] |

Nowadays, different biopolymers, including poly(lactic acid) (PLA), and poly(hydroxyalkanoate)s (PHAs) are producing a capacity growth of approximately 22 %-300 %. These biopolymers offer an eco-friendly pathway to plastic production due to their lower associated carbon emissions from petroleum refinement and extraction [48]. The extensive use of biopolymers has been in composite materials due to their unique attributes, as illustrated in **Figure 1.** These biopolymers act as matrices in composite materials and can be used to replace different thermosets like epoxy resin [49]–[51]. Different methods including in-situ, infiltration, and electrospinning techniques have been used to develop biopolymer composites and have gained considerable attention [52]–[54]. The complete degradation of different biopolymers and their composites is completely achieved in the natural environment due to different micro-organisms, moisture, and ultraviolet (UV) sunlight [55]–[57]. The degradation process performs changes in crystallinity of the biopolymer-based matrix phase and different natural fibers (NFs) or natural residue as reinforcement in composite materials [58].

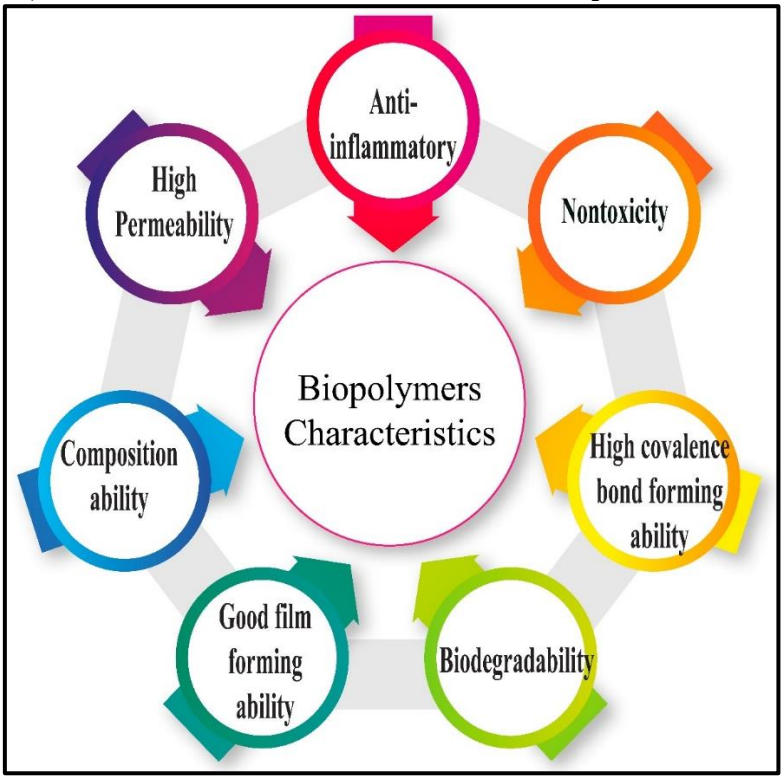



**Figure 1.** Main characteristics of biopolymeric materials.

The world is moving towards a sustainable, cleaner, and greener environment by substituting non-biodegradable polymers with bio-based polymers. These polymers possess distinct

features such as renewability, non-toxicity, sustainability, and bio-degradability [59]–[61]. Therefore, myriad biopolymers are replacing synthetic polymers in different commercial sectors, including packaging, biomedical, and agricultural industries. **Figure 2** presents the major consumption of bio-based polymers in different engineering sectors. The global market indicates that the value of biopolymers increased from $15.6 billion in 2016 to $22.3 billion by 2022.

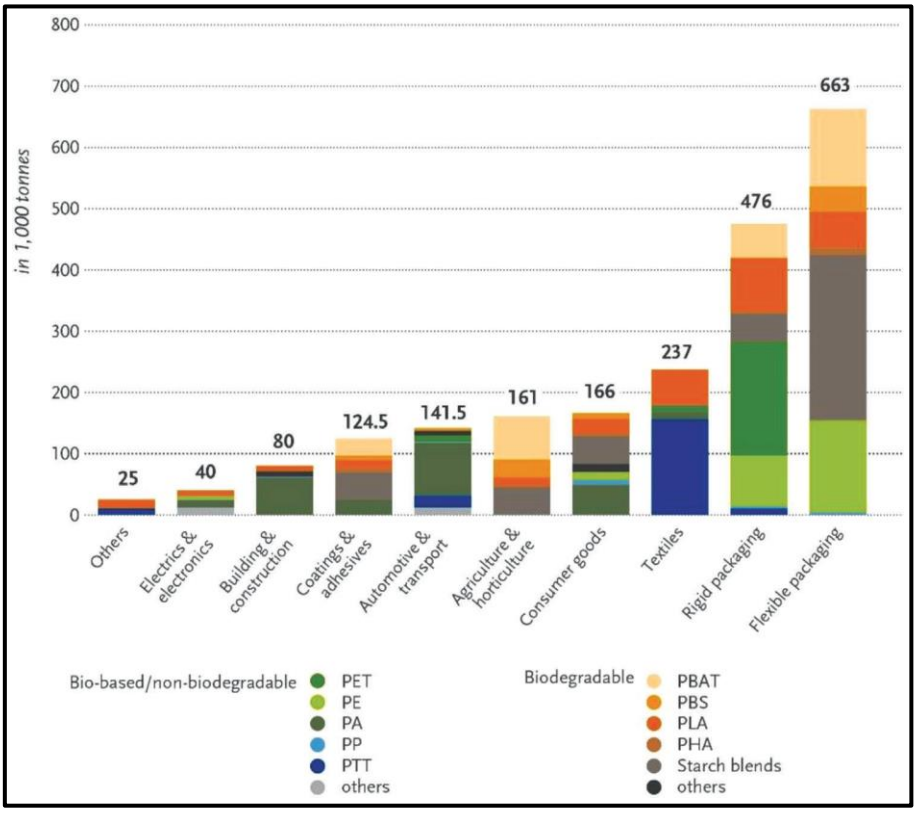



**Figure 2.** Global biopolymer production capacity in different markets **(Adapted from** [62]**)**

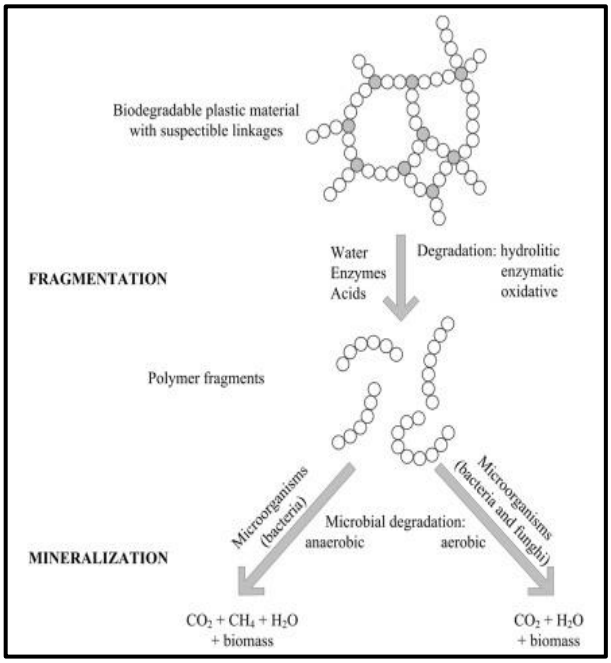
Different natural- and synthetic-based biodegradable polymers have been recently utilized by the research community to evaluate their properties for promoting sustainable solutions. Considering all the aforementioned information about the negative impact of non-biodegradable polymers and positive feedback of biopolymers, the proposed review is written with the aim of highlighting recent developments regarding their biodegradability and the emerging applications of biopolymers in different engineering disciplines.

1. **Biodegradability**

Even the reusing or recycling of non-biodegradable polymers or polymer composites generates harmful and poisonous substances. To overcome this issue, composites are manufactured by using biodegradable-based polymers and fibers, which provide sustainable solutions [63]. Crystallinity and molecular weight are two prime parameters that influence the degradation as well as mechanical properties of biopolymer-based films/coatings. [64]. It is difficult for micro-organisms to attack biopolymers exhibiting molecular weights higher than anti-microbial agents [65]. Thus, the mechanical strength of biopolymers remains unaffected, as anti-microbial molecules do not influence polymeric compatibility. However, any physical or chemical interaction between polymer matrix and additives considerably affects the tensile properties. To overcome this problem, hydrophobic agents, including essential oils, are incorporated as anti-microbial agents to reduce the permeability of the biopolymers. According

to the American Society for Testing of Materials (ASTM), the chemical structure of the biodegradable polymer is significantly transformed by environmental factors. Different enzymes/microorganisms including fungi, bacteria, and algae, deteriorate the chemical and physical properties of biopolymers and reduce their molecular mass by yielding $H_2O$, $CO_2$, and other inorganic products due to anaerobic and aerobic conditions. Additionally, biodegradation produced not toxic or distinguishable residue [66]–[68]. Different degradation strategies, including mechanical degradation, thermo-oxidative degradation, hydrolytic degradation, and photodegradation, also help in promoting biodegradation [69]–[74]. Biodegradation of biopolymers occurs after two steps (depolymerization and mineralization), as elaborated in **Table 2.**

**Table 2.** Chemistry of biodegradation process

| Step | Major stages | Schematic diagram |
|---|---|---|
| Depolymerization | In this stage, macromolecule-based polymeric chains are shortened and weakened under the action of moisture, sunlight, heat, or enzymes [75]. Mostly, exo- or endo-enzymes are abiotic reactions that produce cleavage among the polymer networks. The contact area between biopolymer and micro-organism increases during this stage [76]. |  |
| Mineralization | In this step, transportation of small-sized oligomeric fragments into the cells occurs, where these fragments are bio-assimilation through the microbial population [78]. Depending upon the presence of oxygen, biodegradation is further divided into two types: anaerobic degradation (in the absence of air/oxygen) and aerobic degradation (in the presence of air/oxygen). No residue exists upon the completion of the mineralization process and biopolymer is transformed into biomass, $CO_2$, water, and methane [79]. | Adapted with permission [77] |

### 1.1. Biodegradable Polymers

Biodegradable polymers are obtained from renewable resources and petroleum resources and have recently gained tremendous attraction as these polymers promote a sustainable environment. These polymers are decomposed into minerals, biomass, $CO_2$, $H_2O$, and $CH_4$ through microbial degradation [80]. It is essential to utilize biodegradable polymers to lessen the environmental impact of the climate. On the other hand, biodegradable biopolymers are macromolecular compounds extracted from animals, plants, or micro-organisms and commonly include proteins, lipids, and polysaccharides [81]. Proteins incorporate gelatin, gluten, keratin, and casein, while, lipids include fatty acids or esters, vegetable- and animal-based oils like soya, and sunflower oil [82]. Polysaccharides exist in chitosan, starch, chitin/chitosan, and alginate forms [83].

These biopolymers have drawn significant attention due to their eco-friendliness, non-toxicity, biocompatibility, chemical versatility, and inherent functionality [84]–[86]. On the basis of biodegradability, these polymers are divided into biodegradable and non-biodegradable polymers [87]. Synthetic biodegradable polymer chemical compositions include amides, esters,

diacids, and anhydrides [88]. Repeating units of these polymers are connected through covalent bonds and are fabricated via micro-organisms [89]. Synthetic biopolymers including PLA, PHA, PCL, and PBS are fabricated through chemical processing of different renewable biological sources. Synthetic biopolymers are made up of hydrolyzable links and disintegrate into monomers upon biodegradation [90]. Traditional biodegradable synthetic polymers are synthesized from renewable resources through a hybrid approach that involves chemical and biological reactions [91]. Additionally, valiant efforts have been made for the development of sustainable and green platforms by microbial one-step fermentation of synthetic biopolymers, including PGA, PLA, and multiple types of PHAs [92].

Non-biodegradable biopolymers include polycarbonates, polyethylene, polyether esters, and polyamides (PAs), which are conventionally synthesized from bio-based monomers. Non-biodegradable polymers have been shown to be more usablethan biodegradable biopolymers [93]–[95]. The incorporation of polysaccharide's functional groups into different fillers imparts distinct applications. There are also some non-biodegradable plastics, including bio-PET (bio-ethylene terephthalate), bio-PE (bio-polyethylene), and bio-PTT (bio-polytrimethylene terephthalate) [96]–[98]. **Table 3** summarizes the distinct properties of different natural- and synthetic-based biopolymers, degradation time, and their applications.

**Table 3.** General overview of different biopolymers along with their source, different characteristics, and degradation time

| Biopolymer | Source / composition | Characteristics | Degradation Time | Applications | Ref. |
|---|---|---|---|---|---|
| Collagen | Animal | **(i)** Naturally available protein<br>**(ii)** Biocompatibility<br>**(iii)** Excellent physiochemical properties | 12h | **(i)** Tissue engineering<br>**(ii)** Cosmetic industry<br>**(iii)** Food industry | [99] |
| Alginate | Plant | **(i)** Naturally available polysaccharides<br>**(ii)** Excellent antimicrobial properties | 80 days | **(i)** Textile industry<br>**(ii)** Drug delivery system<br>**(iii)** Tissue engineering<br>**(iv)** Wound dressing | [100] |
| Chitosan | Animal | **(i)** Abundantly available polysaccharides<br>**(ii)** Biocompatibility<br>**(iii)** Excellent solubility<br>**(iv)** Excellent viscosity<br>**(v)** Extraordinary polyelectrolytic behavior<br>**(vi)** Structurally versatile | > 20 weeks | **(i)** Food industry<br>**(ii)** Pharmaceutical industry<br>**(iii)** Agricultural industry | [101] |
| Silk | Animal | **(i)** Naturally available protein<br>**(ii)** Excellent mechanical properties<br>**(iii)** High luster | 6 weeks | **(i)** Tissue engineering<br>**(ii)** Food applications<br>**(iii)** Textile industry | [102] |
| Starch | Plant | **(i)** Naturally available polysaccharides<br>**(ii)** Excellent water vapor properties<br>**(iii)** High viscosity<br>**(iv)** Excellent storage modulus | Several weeks | **(i)** Drug delivery systems<br>**(ii)** Textile industry<br>**(iii)** Paper industry<br>**(iv)** Food packaging | [103] |
| HA | Animal | **(i)** Abundantly available polysaccharides<br>**(ii)** Biocompatibility<br>**(iii)** High viscosity | 4 months | **(i)** Tissue engineering<br>**(ii)** Cosmetic industry<br>**(iii)** Wound dressing<br>**(iv)** Food packaging | [104] |
| Cellulose | Plant fibers | **(i)** Abundantly available in nature<br>**(ii)** Highly crystalline in nature | Weeks to months | **(i)** Biomedical<br>**(ii)** Drug delivery<br>**(iii)** Cosmetics | [105], [106] |

| | | | | | |
|---|---|---|---|---|---|
| | | **(iii)** Good insulation and anti-static properties<br>**(iv)** Hydrophilic in nature | | **(iv)** Clothes<br>**(v)** Food packaging | |
| PTT | Petroleum-based monomers, Produced from 1,3- PDO and PTA/DMT | **(i)** Type of bio-polyester<br>**(ii)** Possess extraordinary mechanical properties<br>**(iii)** Excellent processability<br>**(iv)** Good thermal steadiness | - | **(i)** Textile industry<br>**(ii)** Automotive parts<br>**(iii)** Mobile phone housings | [107] |
| PLA | Corn starch, wheat, sugarcane | **(i)** Very low in price<br>**(ii)** Good resistance to UV radiation<br>**(iii)** Low thermal stability<br>**(iv)** Excellent use as matrix in composite material<br>**(v)** Non-toxic to the environment | 20 months | **(i)** Food packaging<br>**(ii)** Drug delivery<br>**(iii)** Highly used in composite materials | [108] – [110] |
| PHA | Obtained through bacterial fermentation of sugars or lipids | **(i)** Water insoluble<br>**(ii)** Good ultraviolet resistance<br>**(iii)** Soluble in chloroform and other chlorinated hydrocarbons<br>**(iv)** Suitable for many medical applications due to its biocompatibility | 12 months | **(i)** Food Packaging applications<br>**(ii)** Industrial fermentation<br>**(iii)** Animal nutrients<br>**(iv)** Implant material | [111] – [114] |
| PHB | Produced and stored by bacteria | **(i)** Partially crystalline material<br>**(ii)** Stiff and brittle<br>**(iii)** PHB is compatible with the blood and tissues of mammals | 6 to 10 months | **(i)** Wound dressing<br>**(ii)** Act as absorbable nerve guides,<br>**(iii)** Tissue scaffolding bone and nerve regeneration | [115] – [118] |
| PGA | Aliphatic polyester | **(i)** High degree of crystallinity (45–55%),<br>**(ii)** High thermal stability (220–230 °C)<br>**(iii)** High gas barrier (three times higher than ethylene vinyl alcohol) | 5 months | **(i)** Drug delivery,<br>**(ii)** Dental, and orthopedic applications<br>**(iii)** Tissue engineering | [119] |
| PBS | Sugarcane, sugar beet, corn, potato, wheat | **(i)** High heat resistance among the general biodegradability resin<br>**(ii)** PBS has excellent compatibility with a fiber. | 2 to 3 months | **(i)** Bags, food and cosmetic packaging.<br>**(ii)** Disposable products<br>**(iii)** Agriculture industries | [55] |
| PVAc | Polyvinyl acetate | **(i)** Nontoxic and thermoplastic polymer<br>**(ii)** Partially crystalline<br>**(iii)** High tensile strength and flexibility<br>**(iv)** Soluble in water | Several months to years | **(i)** Textile and paper industry<br>**(ii)** Food packaging<br>**(iii)** Biomedical applications | [120] |
| PCL | Petroleum-based products | **(i)** Excellent chemical and solvent resistance<br>**(ii)** High employed in composite material<br>**(iii)** Highly elastic in nature | 6 to 28 months | **(i)** Surgical implants<br>**(ii)** Drug delivery devices<br>**(iii)** Regenerative medicine | [121] – [123] |

### 1.1.1 Biodegradation mechanisms

Biodegradation of biopolymers occurs through the action of microbial microorganisms including algae, fungi, or bacteria. For instance, Kalita et al. [124] evaluated the biodegradation mechanism of algae/PLA-based composites and compared their performance with PLA-based composites. **Figure 3($a_1$)** depicts steps involved in the biodegradation of biopolymer composite. ASTM standard D5338–15 was employed to evaluate the degradation behavior

under thermophilic composting and abiotic conditions. Scanning electron microscope (SEM) results provided in **Figure 3($a_2$)** showed that rough patches containing holes and cracks were developed in biopolymer due to the excellent biodegradation. Additionally, PLA/ 5% algae extruded sheets (ES) specimens exhibited higher microbial attachment after two weeks of biodegradation, as illustrated in **Figure 3($a_3$).** The excellent degradation of biopolymers occurred due to the presence of high nitrogen content in biomass (algae).

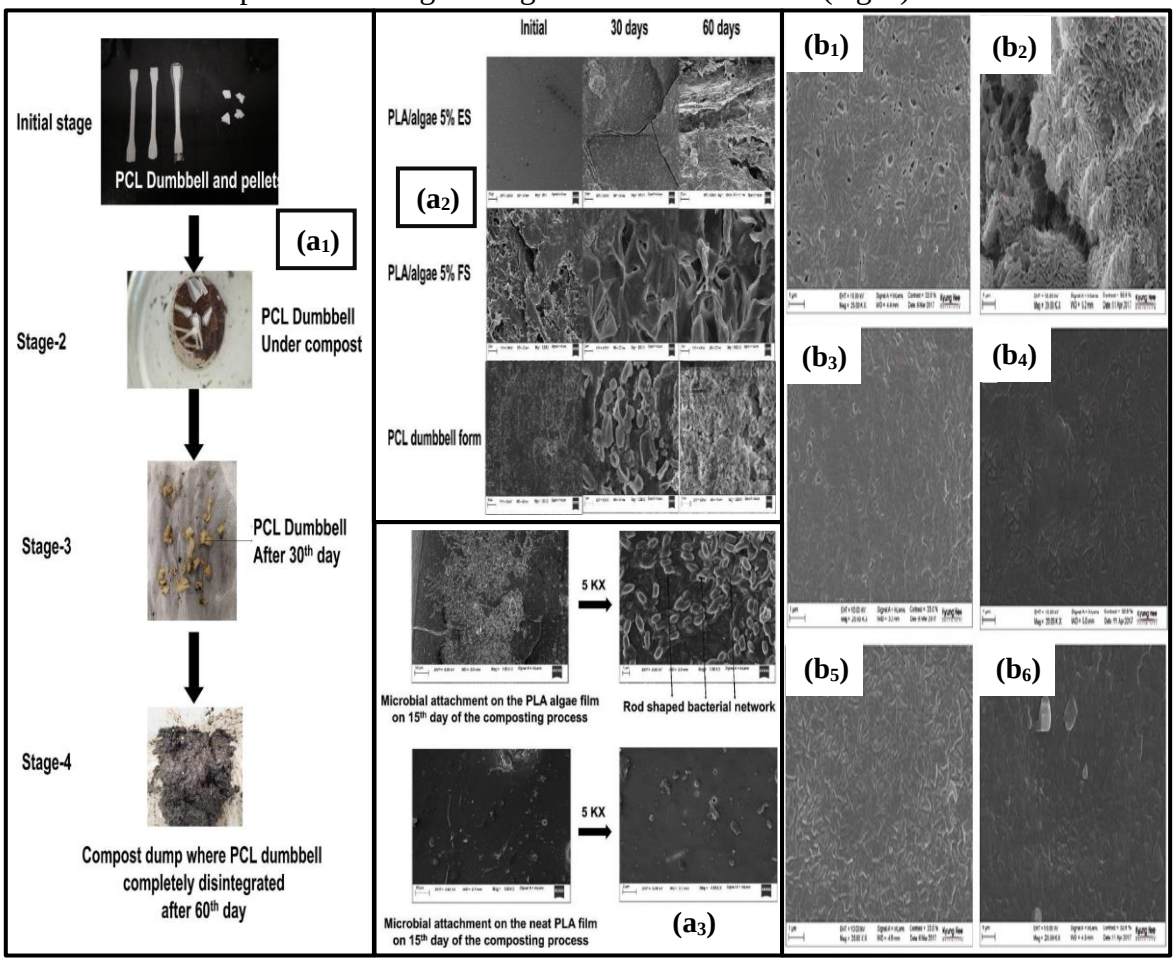



**Figure 3.** ($a_1$) Flowchart showing different stages of the biodegradation mechanism of PCL-based biopolymer; ($a_2$) SEM images of the biodegraded PLA-based polymers; ($a_3$) Microbial attachment comparison on the PLA/5 % algae ES and neat PLA ES films on the fifteen day of the composting process. (Adapted from [124]); SEM images depicting ungraded and degraded samples; ($b_1$, $b_2$) PEO40/PLA60/CNT1-based biopolymer before and after four weeks degradation; ($b_3$, $b_4$) PEO25/PLA75/CNT1-based biopolymer before and after four weeks degradation; ($b_5$, $b_6$) PEO10/ PLA90/CNT1-based biopolymer before and after four weeks degradation [125] **(adapted with permission)**

The combination of inorganic nanomaterials, including nanowires, carbon nanotubes (CNTs), carbon nanofibers, or graphene, has opened a new research field, which helps in developing high-performance nano-devices [126]–[128]. For example, Zare et al. [125] developed poly(ethylene oxide)(PEO)/PLA-based nano biosensor by incorporating CNTs. SEM results observed for this study are shown in **Figure 3(b**) which demonstrated that the addition of CNTs accelerated degradation in PLA/PEO-based biopolymers by dwindling the interphase thickness during degradation.

Similarly, Urbanek et al. [2] proposed a low temperature biodegradable study of PCL, PBS, and PBSA using Antarctic microorganisms, as illustrated in **Figure 4(a)**. The reported results confirmed that highest biodegradations were achieved in the order of 49.68% for PBSA and 33.7% for PCL, 45.99% for PBSA, and 49.65% for PCL, respectively at 20°C under antarctic microorganisms, as depicted in **Figure 4(b)**. Finally, these antarctic microorangisms are best suited for effective plastic degradation at relatively low temperatures.

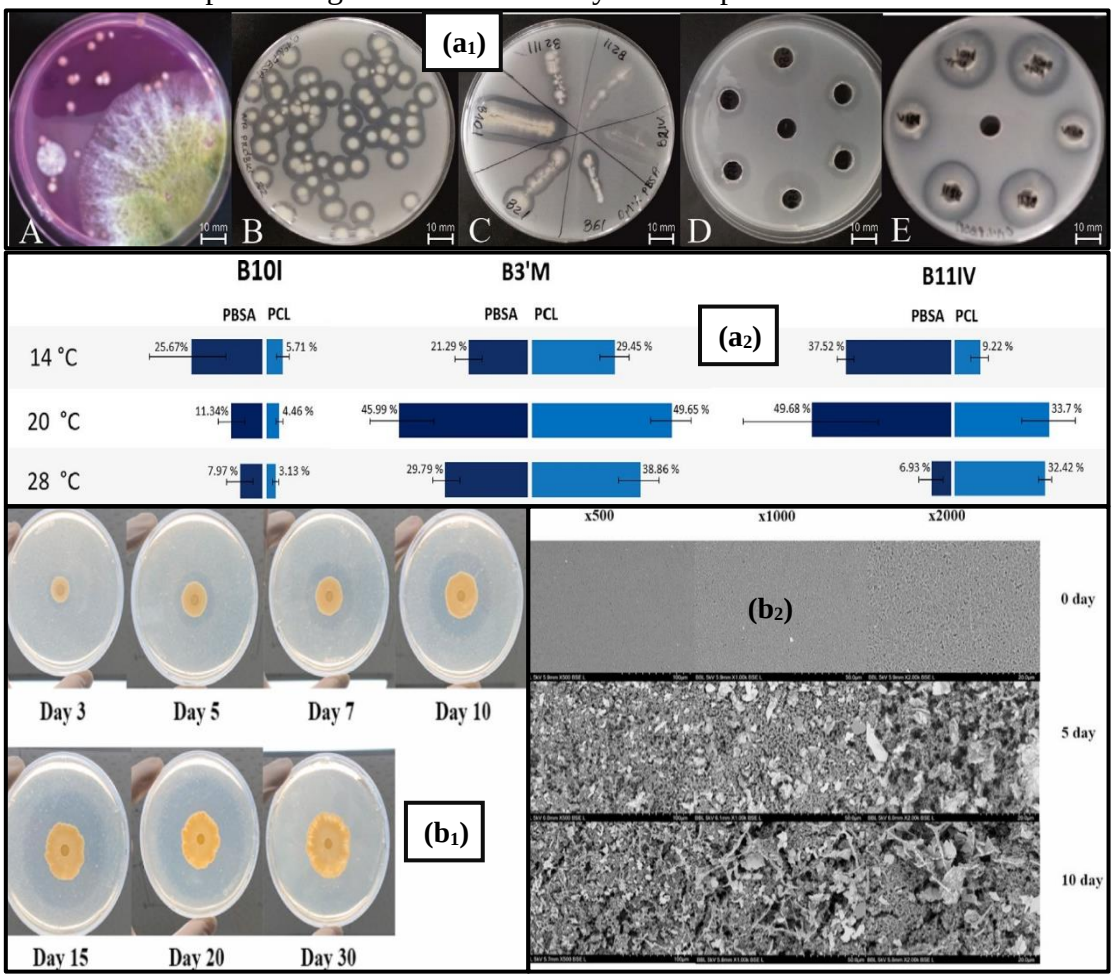



**Figure 4. ($a_1$)** Isolated antarctic microorganisms and halo zones that appeared around the colonies on FMM medium supplemented with 0.1 % PBSA (B, C, D, E) showing biodegradation; ($a_2$) Biodegradation rates for different studied biopolymers at different temperatures [2] (Adapted with permission); ($b_1$) Clear zones images representation generated by Bacillus sp. JY14; ($b_2$) SEM images depicting the surface changes as a result of degradation [129] (adapted with permission)

Cho et al. [129] evaluated different properties of PHB-based biopolymer through six different microorganisms commonly found in marine soil at different temperature and salinity levels. The PHB-degradation was monitored by noting the changes in the physical and chemical properties of PHB films incubated with Bacillus sp. JY14, as depicted in **Figure 4($b_1$). SEM results provided in Figure 4($b_2$)** indicated transformation in the surface structure, molecular network, and weight of PHB-based biopolymer upon degradation. Additionally, a maximum of 98 % PHB degradation was achieved in just 14 days.

Similarly, Ruggero et al. [130] performed lab-scale degradation testing on Mater-Bi® (MB) film starch-based biopolymer and PBAT under different temperature and moisture conditions. **Figure 5(a)** depicts SEM images of degradation behavior in which circular spots disappeared and formed small holes. The results demonstrated that the biodegradation of PBAT is highly

affected by temperature and moisture conditions. Furthermore, among all three replicates, both starch and additives are completely decomposed within the first few days of the degradation process.

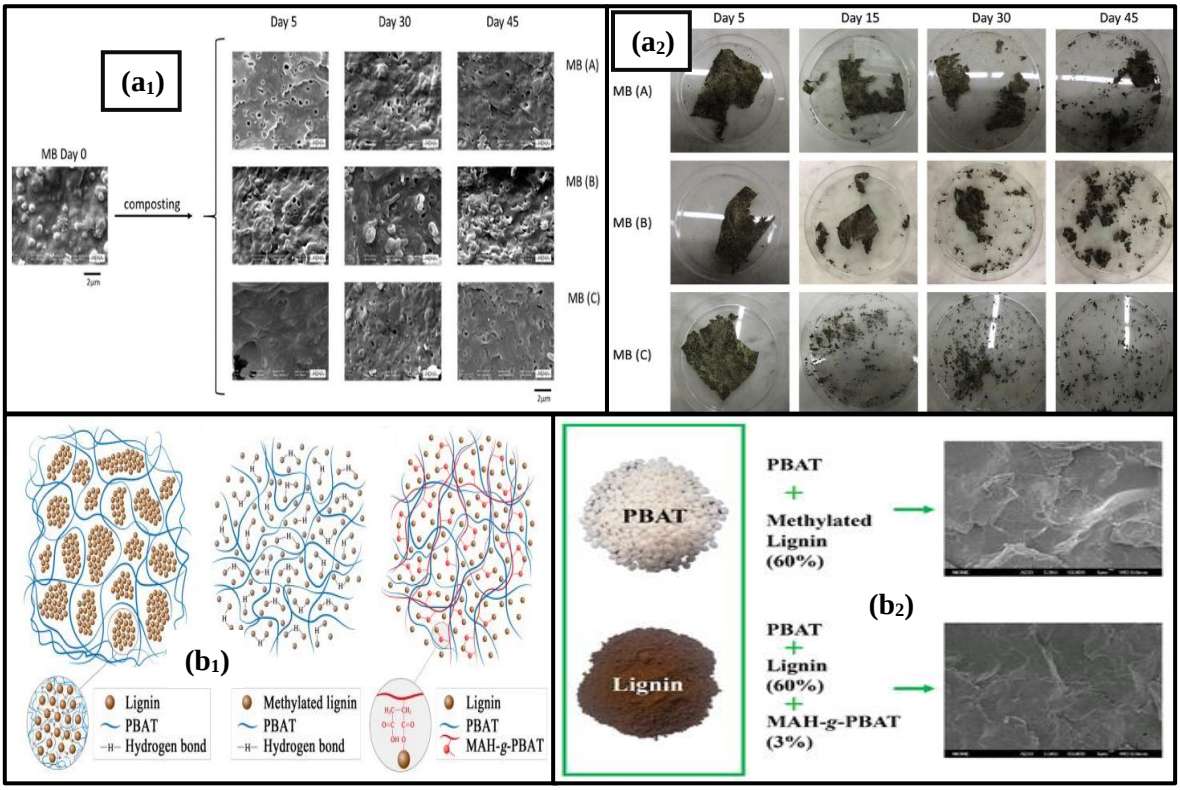



**Figure 5.** ($a_1$) SEM images of different A, B and C degraded samples during 5, 30, and 45 days of the composting process; ($a_2$) SEM images of the degraded samples during 5, 15, 30, and 45 days of the composting process [130] (Adapted with permission); ($b_1$) Lignin/PBAT-based biodegradable composite films/materials (Adapted with permission from [131]).

Wang et al. [132] developed lignin-based biodegradable composites by incorporating lignin into the PBAT matrix. For improving the compatibility, a green esterification reaction under microwave-assisted solvent-free conditions was adopted. These cost-efficient biodegradable materials with controlled mechanical properties are best suited for UV-shielding properties for packaging applications. In another study, Xiong et al. [131] evaluated the effects of lignin methylation and compatibilizer on biodegradable PBAT/lignin composites produced through melt extrusion, as depicted in **Figure 5(b)**. It was reported that the degree of lignin aggregation played a crucial role in the mechanical properties of these biodegradable composites. These biopolymer-based composites have a strong potential to be manufactured into rubbish bags, packaging bags, and soil remediation films. To summarize this, biopolymers exhibit excellent biodegradation behavior and comparable mechanical properties. These polymers can be composted leaving environment-friendly by-products including $CO_2$ and water.

### 1.2. **Biodegradable composites**

Biocomposites are environmentally benign, sustainable, renewable, carbon-neutral materials and eco-composites, which use NFs and biopolymers. In the last decade, these materials have spellbound the world due to their lightweight nature, recyclability, and cost-effectiveness [133]. Additionally, these composites contain fillers/additives and bio-based polymers that originate from renewable sources [134]. The decomposition of biocomposites occurs by soil burial or outdoor climate. Degradation occurs due to the complete breakdown of NFs that

damages the bonding between the fibers and biopolymer matrix. The biodegradability of biocomposites is influenced by UV light, temperature, air dampness, and microorganisms [135].

Nowadays, in biopolymeric composites, different NFs including plant-based (jute, cotton, bamboo, and flax), animal-based (silk and wood), mineral-based (asbestos), and microbial-based (polycyclic aromatic hydrocarbons, and bacterial cellulose), are vastly applied as reinforcement materials [136]. These fibers promote a sustainable green environment due to their appropriate mechanical characteristics and availability [137]. Because both fibres (reinforcement) and biopolymers (matrix) are derived from renewable sources and are compostable, biocomposites are excellent substitutes for carbon- and glass-reinforced polymer composites [138]. For instance, Duigou et al. [139] evaluated the mechanical performance of PLA/flax-based composites which exhibited mechanical properties comparable to polypropylene (PP)/hemp, PP/glass, and PP/sisal composites. Additionally, the shortening of fibers and separation of fiber bundles due to the recycling of composites did not influence the mechanical properties. Hence, these biocomposites are promising candidates for promoting a green and eco-friendly environment.

Biocomposites are mostly manufactured through the electrospinning technique which exhibits the versatility to use incorporate/reinforce continuous nano-fibers (2 – 500 nm) into different biopolymers, compared to other nano-fiber processing routes [140]. Additionally, this process possesses other advantages including tailorable physical properties, fiber functionalization, deposition upon substrate, material combination, and mass production capability [141]–[143]. Electrospun manufactured composites show high micro-sized pores and high surface area to volume ratio making them suitable for biomedical and air-filtration applications [144].

Similarly, the reinforcement of NFs in biocomposites as additives or fillers can be done through fused filament fabrication (FFF). The properties of the 3D-printed biocomposites depend upon the type of fibers, their strength, stiffness, and interfacial bonding with the biopolymer matrix [145]. **Figure 6** depicts different sustainable reinforcements and fillers which are employed for the fabrication of biocomposite materials [146]. The primary purpose of these reinforcements is to reuse different recyclable materials [147]. It lowers manufacturing costs while increasing biodegradability in biocomposites [148]–[150].

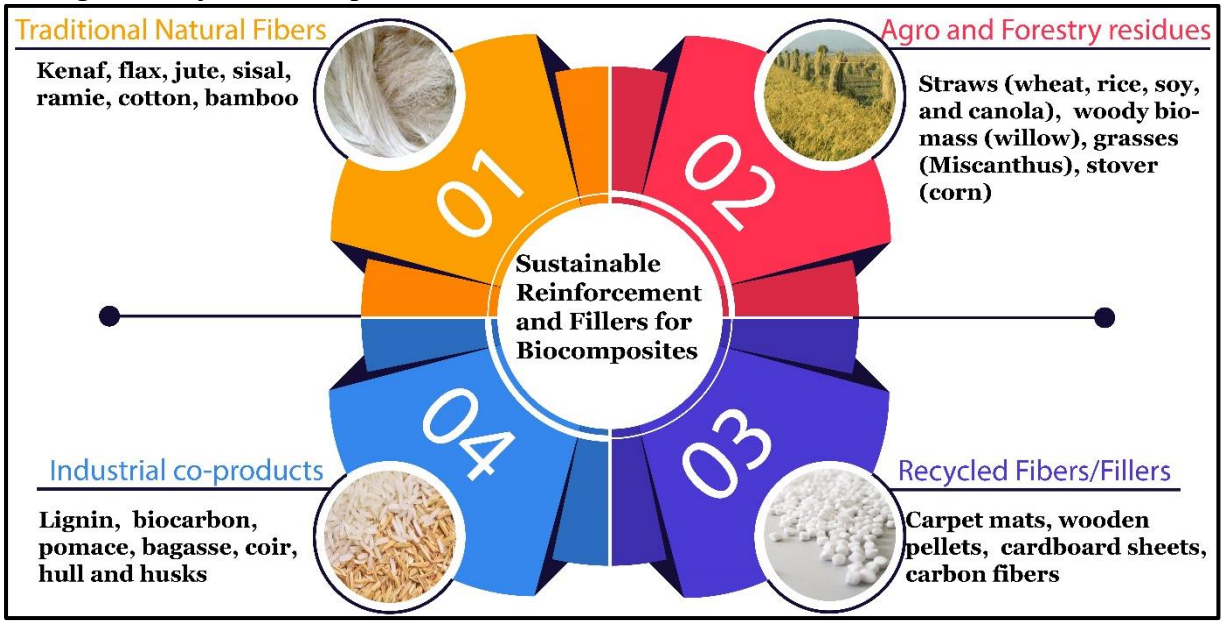



**Figure 6.** Different types of natural fibers employed for the manufacturing of biocomposites

The infusion of bio-derived nanofibers and nanoparticles (NPs) significantly improves mechanical and physical properties, including heat deformation temperature, modulus, gas permeability, tensile behavior, and decomposition of the developed composites. Most biocomposites contain starch, cellulose, PLA, PCL, PBS, PVA, and polyhydroxy butyrate (PBH). For instance, the reinforcement of sugar palm fiber in the biopolymeric composite significantly improved the mechanical characteristics [151]. The matrix constituent of the biocomposites is responsible for imparting better mechanical features with regard to their morphology, chemical constituents, molecular weight, and processing approach.

Different biodegradable fiber/polymer-based composites are evaluated by using various ASTM standards. For instance, soil burial testing calculates biodegradation by assessing the mass loss of polymers/fibers after testing [152]–[154]. Additionally, soil degradation occurs upon exposure to soil microbiomes. Soil burial testing is performed as per ISO 17556 and ASTM D5988-18 standards. Different influencing parameters, including moisture-holding limit, damping content, pH level, ash content, and carbon-to-nitrogen ratio have also been encountered during testing. Burial period and damping content have considerably influenced biodegradability, out of all these parameters [155]–[157]. **Table 4** incorporates the most recent studies of biodegradable NFs- and biopolymer-based composites, their testing techniques, degradation percentage, and time.

**Table 4.** Compilation of the recent work on the biodegradability of different NFs- and biopolymer-based composites

| **Biodegradable fiber- and polymer-based composites** | **Source** | **Biodegradation** | **Degradation time** | **Testing method** | **Ref.** |
|---|---|---|---|---|---|
| CA | Wood Pulp | DS* of 1.8 achieved | Varies from several days to week | Reaction of CA with biocatalysts | [158] |
| PBS/jute composites | Cellulose | 62.5 % | 180 days | Soil burial | [159] |
| PLA | Corn, starch, sugar | 100 % | 9-12 days | Incubator with *Burkholderia cepacia* composite | [160] |
| Sisal fiber and TPS | Agave sisalana, starch | 62 % | 28 days | Soil burial | [161] |
| OPEFB fiber/ACC films | Palm oil plant | 56.56 % | 28 days | Soil burial | [162] |
| Polyester–Banana Fiber | Banana/euphorbia coagulum | 41 % | 90 days | ASTM D 5988 | [163] |
| PLA-grafted-MA/GCF | Corn, starch, sugar | 92 % | 21 days | Incubator with *Burkholderia cepacia* compost | [160] |
| PE/NR composite | NR | 38.3 % | 90 days | Full-scale soil test (ASTM D 5988) | [164] |
| TPS composite | Starch | 88.57 % | 30 days | - | [165] |
| Feathers nonwoven reinforced polyester composite | Chicken feather fiber | 62 % | 60 days | Soil burial period | [166] |
| BF-reinforced tapioca starch PVA composite | Starch, cellulose | 27 % | 15 days | ASTM G160 | [167] |
| Garden waste cellulose in starch/PVA | Starch, cellulose | 80-91 % | 14 days | Soil burial | [168] |
| Rich husk filler PLA composite | Rice husk, PLA | 3.3 % | 1 day | ASTM D-5488-94d | [169] |
| MOSF based PVA composites | Crude protein, crude oil | 5-35 % | 90 days | Soil burial | [170] |

| | | | | | |
|---|---|---|---|---|---|
| Corn based SB and CB, Cassava based SB and CB | Starch | 12-57 % | 14 days | Soil burial | [30] |
| Starch/MC composite | Starch, banana stem | 82-96 % | 28 days | Soil burial | [171] |
| SPNCC-based nanocomposites and SPS films | Cellulose, Starch | 100 % | 9-12 days | Soil burial | [172] |
| Starch/Chitosan/PVA composite | Chitosan, Starch | 90 % | 90 days | ISO 846:1997 | [173] |
| PBS/levan composite | Polyester | Holes due to biodegradation were visible | 4 days | Soil burial | [174] |
| RS-g-PMMA bio composite | Lignocellulosic | 11 % | 15 days | APHAS method | [175] |
| MNP/PVA Composite | MNP | 0.141, 0.046, and 0.027 mg | Per hour | *Zophobas morios* Larvae (Superworms) | [176] |
| Ramie-based PLA composite | PLA | 100 % | 60 days | Soil burial | [154] |
| CNC/chitosan composite | Cellulose | Moderate biodegradability | 15 days | Soil burial | [177] |
| WPC | Cellulose | 3.55 weight loss per gain | - | ASTM D5338-11 | [178] |
| BF/PLA composite | Lignocellulosic, PLA | 12-28 % | 10-70 days | ASTM D5338-11 | [179] |
| AEMO-NVP composite | Leather | 1.25-5 % | 7-28 days | Soil burial | [180] |
| Oxo-PP composite | Transition metal ions | 28.87 % | 45 days | ASTM D 5338 | [181] |
| PWBF-PVA composite | Cellulose, PVA | 3.5 g weight loss was observed | 60 days | Soil burial | [182] |
| Jute composite | Cellulose, rice bran | 0.2-2.91% | 7-28 days | Soil burial | [183] |
| Polyester composite | Shorea robusta | 6.2 % | 15-120 days | Soil burial | [184] |
| FCC | Cellulose | 48 NI $kg^{-1}$ net biogas produced as a result of biodegradation. | 21 days | German standard DIN 38414 | [185] |
| rPP and KF composites | Cellulose | 11.82 % | 4 months | Soil burial | [186] |
| Coffee wastewater | Carbohydrates | 80-90 % BOD | 10-60 days | Simulation technique | [187] |
| CG and gelatin films | Gelatin | 45 % | 22 days | ASTM D5988-12 | [53] |

*****DS: Another way of measuring biodegradability. A level of 1.8 DS shows good biodegradability.

In summary, biopolymeric composites are sustainable and eco-friendly materials, which exhibit excellent biodegradability and mechanical properties. These composites promote a cleaner and greener environment. The development of composites by using recycled fibers gives comparable mechanical properties and can be applied for high-performance applications.

## 3. Life cycle of biopolymers

Life cycle assessment (LCA) is a system employed for analyzing the environmental and ecological footprints of products and materials. This is usually done through the evaluation of different life stages of products, including extraction of raw materials, fabrication, use, and end-of-life stage [188]. The ultimate life cycle of biodegradable materials is depicted in **Figure 7.** In the first step, biopolymers are often derived from agricultural products and involve the cultivation of different raw materials, such as potatoes for starch, wood for cellulose, or any crop used. These polymers are manufactured in substages: fertilizers, pesticides, and herbicides

also help in the manufacturing of biopolymers. Production and milling are the other two key processes, which help in developing biopolymers [189]. In the second step, these polymers are employed to develop different consumer products which exhibit improved physical and mechanical properties. Additionally, the mechanical properties of these composites can be enhanced by infusing organic nano-materials [190]. In the third step, the EOL of biopolymer-based consumer products seeks to evaluate the impacts associated with the production and disposal of biopolymers compared to fossil-based plastics. Different routes, including landfill, compost, and recycling, are used for different biopolymers, whereas some compostable biopolymers including PLA and TPS are modeled using compost and landfill scenarios [191]. In the last step, biopolymer degradation occurs in the biological environment through different microorganisms including algae, fungi, and bacteria [192]. Consequently, more than 90 % of biodegradable polymers are transformed into water, $CO_2$, and minerals within six months as per EU standard.

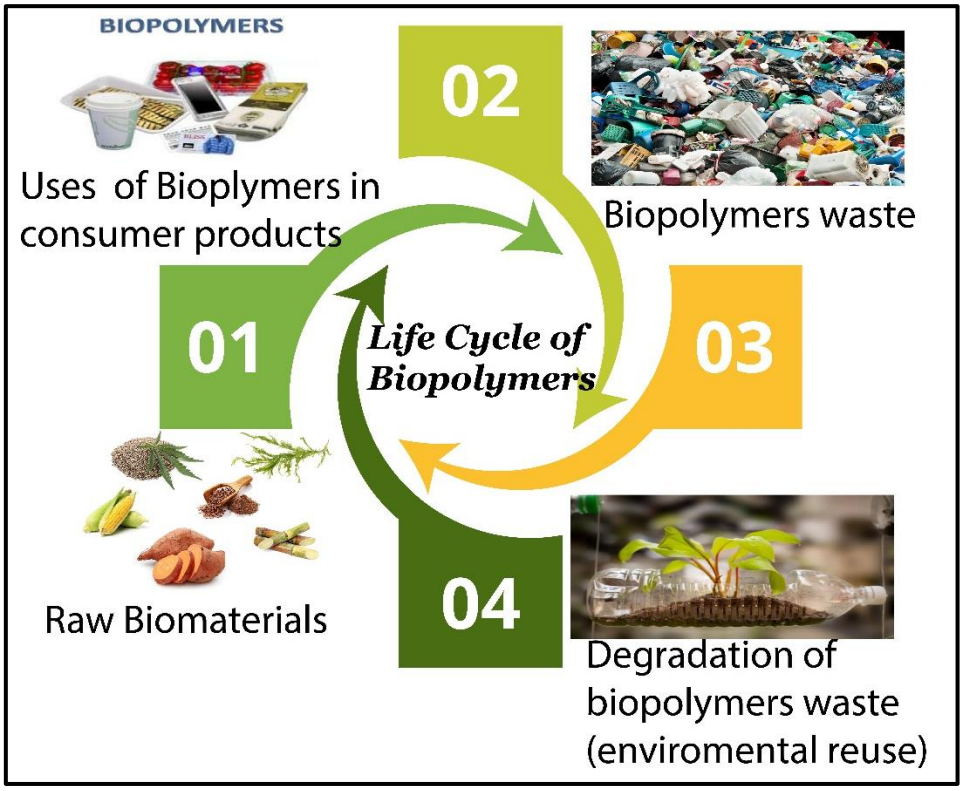



**Figure 7.** Life cycle of biopolymers.

4. **Applications of Biopolymers**

Biopolymers have exhibited huge potential due to their flexibility in chemistry that gives rise to materials with a great diversity of physical and mechanical properties [193]–[195]. Since their emergence, biopolymers have been effectively utilized in many biomedical and other engineering applications including controlled drug delivery, food packaging, construction industry, regenerative medicine, wearable electronics, orthopedic, and long-term implants [196]–[198]. Rapid advancement in materials science and technologies have opened up new windows of opportunities to discover the novel applications of biopolymers in engineering and biomedical fields [199]–[201]. The focused researches on these materials are reflected by a significant upsurge of the biodegradable polymer-based marketed products and ongoing clinical trials of these materials [202]–[204]. Particularly, in the last decade, biopolymers

experienced a renaissance due to the limitation of fossil resources in combination with public demands for environmentally-friendly and sustainable processes, which has led to the formation of a market for bio-based plastics [205].

These eco-friendly biopolymers and their composites can be classified based on their various sources, numerous techniques of manufacturing, and possible forms of usage [206]. Because of their excellent biocompatibility and biodegradability, these polymers are ideal for use in environmental applications [207]–[209]. Biopolymer-based materials are also responsible for reduced carbon footprint during their manufacturing and their microbial degradation improved overall resource efficiency [210].

### 4.1 **Medical applications**

Numerous biodegradable polymers and composites including PLA, PVA, PCL, and PHB are being used in tissue engineering [211], orthopedic implants [212], and drug delivery [213]. These biodegradable materials have no harmful impact on the human body, thus, making them highly appropriate for medical applications [214]. PCL-based materials have a wide scope in wound healing applications, drug delivery systems, and tissue engineering applications, especially in the formation of hard tissues, soft tissues, and load-bearing tissues [215]–[217]. **Table 5** contains some of the most recent studies containing the applications of biodegradable biopolymers and their composites.

**Table 5.** Recently adopted different biopolymers based single/hybrid materials and their engineering/biomedical applications.

| **Researcher** | **Year** | **Biopolymers** | **Added materials / fillers** | **Engineering applications** |
|---|---|---|---|---|
| Zare et al. [125] | 2022 | PLA/PEO | CNT | Degradation-based nanobiosensor |
| Yang et al. [218] | 2020 | PLLA | Xeolitic imidazolate framework-8 (ZIF-8), | Bone scaffold |
| Yuan et al. [219] | 2019 | Bacterial cellulose and chitosan | Collagen | Hemostatic material |
| Mishra et al.[220] | 2021 | Chitosan-casein | Nanofibrous polyelectrolyte complex | Hemorrhage treatment, provide rapid blood clotting |
| Yalcin et al.[221] | 2021 | PHB | siRNAs | Gene delivery system |
| Malik et al. [222] | 2018 | PCL | Organo modified montmorillonite clay | Fixation devices |
| Mucke et al.[223] | 2021 | TPS | PBAT | Packaging |
| Kundu et al. [224] | 2021 | PCL | nHAP fibers | Tissue engineering |
| Diaz et al. [225] | 2020 | PCL | nHAP/MWCNT | Bone regeneration |
| Villamagna et al. [226] | 2020 | PEA | Peroxisome proliferator-activated receptor δ | Drug delivery |
| Das et al. [227] | 2019 | PLA | MWCNTs | Chemo, photothermal therapy and tissue engineering |
| Mondal et al. [228] | 2020 | PLA | HAP | Drug delivery and tissue engineering |
| Singh et al. [229] | 2019 | Chitosan | Nano-sized BG | Bone scaffold and bone tissue regeneration |
| Lucik et al. [230] | 2020 | PLA/PCL | Thymol and carvacrol mixture | Food packaging, |
| Mouro et al. [231] | 2021 | PCL/PVA | Chelidoniummajus L. | Wound dressing |
| Chugh et al. [232] | 2020 | PEA | Dimer acid | Hot-melt adhesive |

| | | | | |
|---|---|---|---|---|
| Tseng et al. [233] | 2020 | PLA | - | cutlery products |
| Zhu et al. [234] | 2020 | PVA/CMC | $Na_2CO_3$ | Paper processing, adhesives and coatings, water-soluble films, emulsifiers, textiles |
| Ge et al. [235] | 2021 | Gelatin | Agarose gel | Gelling agent |
| Mirzaeei et al. [236] | 2021 | PCL and PLGA | Metronidazole and amoxicillin | Drug-delivery systems |
| Dietrich et al. [237] | 2019 | PHB | Hardwood holocellulose hydrolysate | Detoxification system |
| Zhao et al. [238] | 2020 | Chitosan | EDC and NHS | Artificial liver |
| Zhao et al. [239] | 2019 | PLGA | MSCs | Ligament/Tendon Repair |
| Buyuksungur et al. [240] | 2021 | PCL | GelMA | Dentistry |
| Parvizifard and Karbasi [241] | 2019 | PHB | MWCNT | Tissue engineering |
| Ferlic et al. [242] | 2020 | PHB | $ZrO_2$ | Surgical implant |
| Deng et al. [243] | 2021 | PCL | PEG/chitosan-keratin | Sutures |
| Arjona et al. [244] | 2021 | PHB | - | Microcapsules |
| Subramanian et al. [245] | 2020 | PCL | SMA hydrogel | Contraceptive devices |
| Caballero et al. [246] | 2021 | Alginate | - | Theragnostics |
| Robles et al. [247] | 2019 | PLA | Lignin | Wound healing |

Besides these, biopolymers are also promising materials for many filter applications due to their desirable chemical and physical properties which are best suited for absorption and the removal of certain chemical contaminants, stipulate bactericidal, or viricidal functionality. Different functional groups present in proteins and polysaccharides permit highly selective filtration for pollutants and other contaminants [248], as illustrated in **Figure 8(a)**. Hemorrhage or excessive blood loss during childbirth, accidents, and complex surgeries is the leading cause of death. Bleeding management is an effective strategy to control excessive blood loss, which can be achieved using topical hemostatic agents through different biopolymers [249].

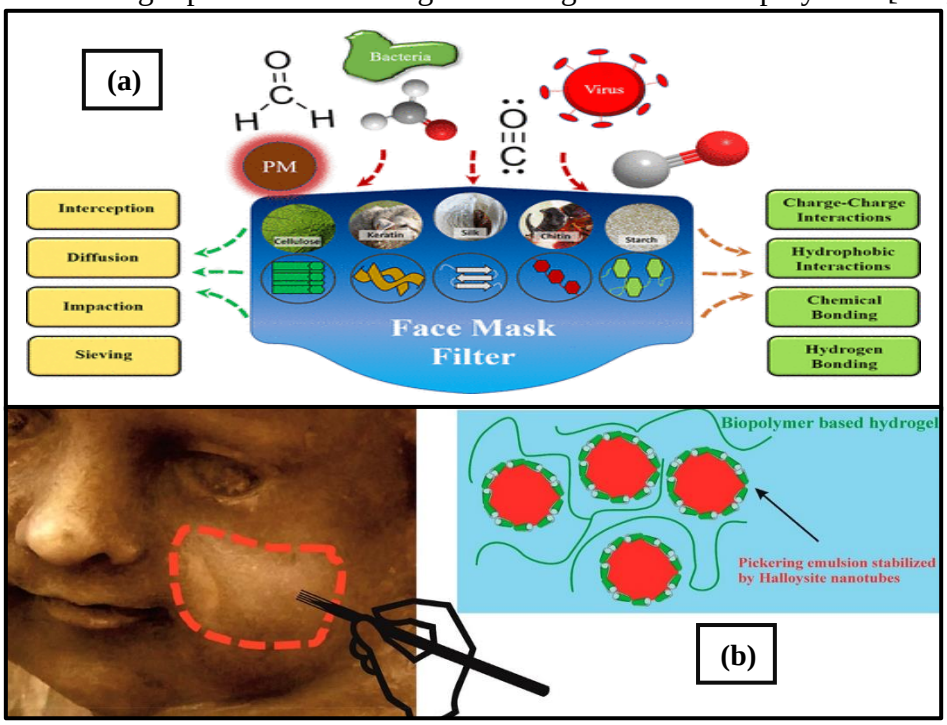


**Figure 8. (**a) Filtration materials produced from different biopolymer including variety of protein cellulose, silk, keratin, chitin, and starch [248] (adapted with permission); (b) Pickering emulsions based on halloysite nanotubes and ionic biopolymers (chitosan and pectin) [250] (adapted with permission).

Cavallaro et al. [250] fabricated sustainable Pickering emulsions using halloysite nanotubes and ionic biopolymers including chitosan and pectin. The reported results showed that Pickering emulsion in a pectin-based gel phase has the potential for wax layer removal from a marble surface, as depicted in **Figure 8(b)**. Furthermore, pectin improves the Pickering emulsion stability, while a nonhomogeneous gel and phase separation were detected in the presence of chitosan.

### 4.1.1 **Tissue Engineering**

Tissue engineering is a reliable method for supplying perfectly matched organs for the necessary transplantation of the patient due to injury, trauma, or any chronic disease [251]. Thus, tissue engineering is a lifesaving technique due to the shortage of donors at vital times when a patient's life can be in severe danger [252]–[255]. To date, different organs/parts including kidney, liver, orthopedic, nerve, pancreas, ear, and nose have been implanted. This is entirely possible due to scaffolds fabrication with the use of 3D-printed biodegradable polymers [256]–[259]. These biopolymer-based printed scaffolds have excellent biodegradability, compatibility, porosity, osteointegration, size, and bioactivity [193], [260]–[262].

For instance, Jinga et al. [263] developed composite scaffolds by using PCL-based biodegradable polymer along with other inorganic mineral powders including zinc oxide (ZnO), titanium dioxide ($TiO_2$), and hydroxyapatite (HAP) via electrospinning method. Upon successful experimentations, it was noted that these fiber-based composites are accepted by cell structures. Hence, these scaffolds could be used for regeneration of damaged parts and wound healing applications. The PCL/collagen-based electrospun fibrous membrane was evaluated for wound healing applications through an in-vitro study [264], as depicted in **Figure 9(a)**. The results indicated that biopolymer-based exhibited polarized and ordered fibroblasts, and these fibroblasts possessed excellent cell proliferation and focal adhesion. A randomized controlled clinical trial targeted to estimate the feasibility and effectiveness of PCL-based scaffold in fresh extraction sockets for ridge preservation was employed [265], as depicted in **Figure 9(c).**

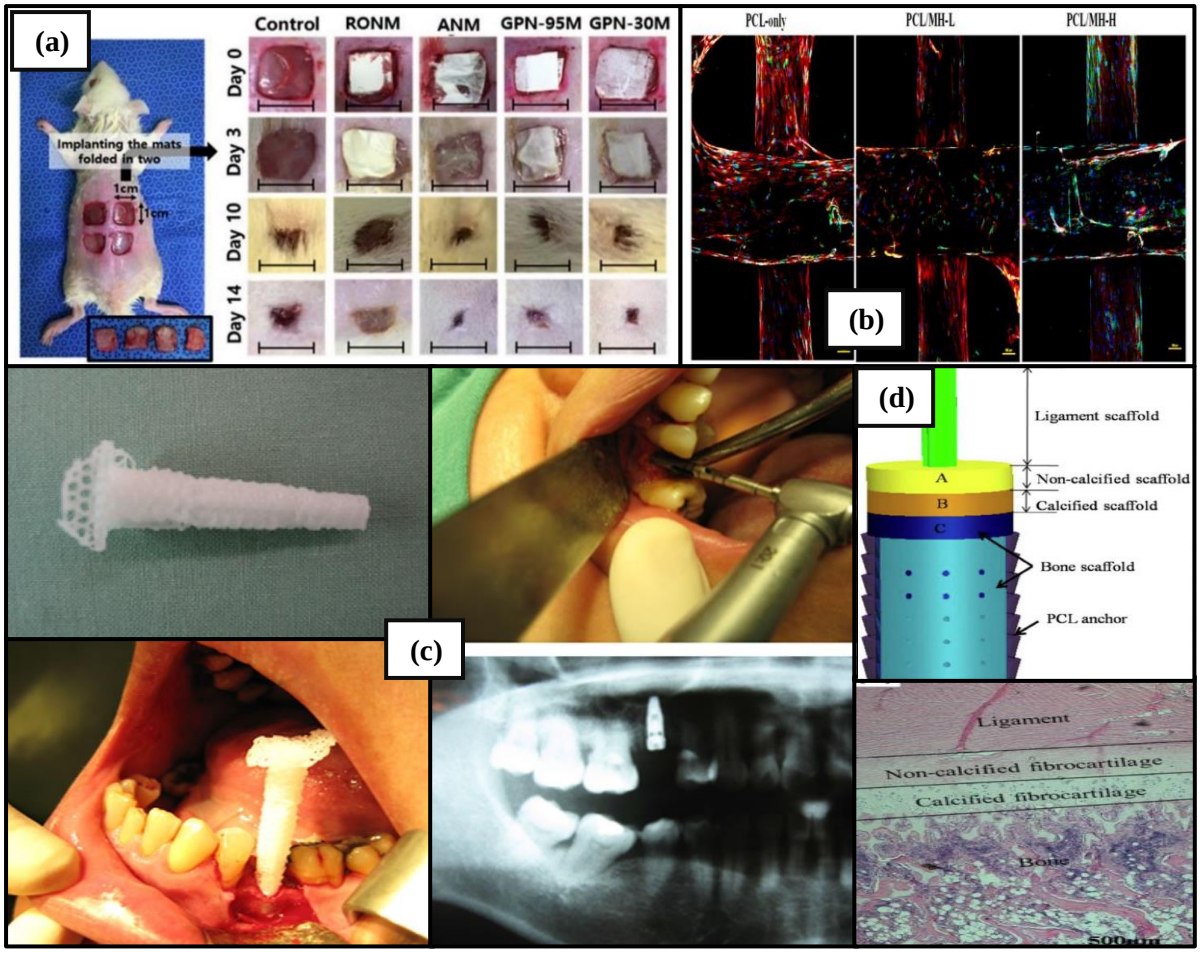



**Figure 9**. **(a)** Time dependent healing process of rat wound using PCL/collagen mats [264] (Reproduced with permission); (b) Immunostaining for the osteogenic marker images of osteoblasts on 3D-printed scaffolds (Adapted with permission from [266]); (c) PCL scaffold during surgical implant and tooth extraction site [265] (Reproduced with permission); (d) Design of a scaffold for ligament, cartilage, and bone generation [267] (Reproduced with permission)

Similarly, PCL/HAP/simvastatin electrospun nanofiber-based composite coating was deposited on a magnesium substrate by Rezk et al. [212] and observed to have excellent biodegradability and enhanced mechanical properties, which could be helpful for their utilization in orthopedic implants. In another study, Abdal-hay et al. [266] 3D-printed a PCL-based biodegradable composite by incorporating bioresorbable magnesium hydroxide (MH) NPs. The results revealed that the inclusion of MH significantly improved the mechanical properties and accelerated the degradation rate of PCL in phosphate-buffered saline solution. Furthermore, the cell bridging was greater on the PCL/MH of 20 wt.% compared to PCL- and PCL/MH (5 wt.%) scaffolds. Additionally, full bridging of scaffold pores was noted in the PCL/MH of 20 wt.% scaffolds. The cell structure and bioactivity were also examined using immunofluorescence staining, as shown in **Figure 9(b).** Thus, these materials have broad implications for the manufacturing of bioactive 3D-printed scaffolds with tailorable degradation properties and are ideal candidates for the regenerative treatment of various injuries or bone diseases.

Soni et al. [268] proposed a promising technique for the healing of bone tissues using biocompatible and biodegradable bioactive glass/PCL-based thin membranes and observed excellent mechanical properties and degradation. **Figure 9(d)** illustrates the multiphase

scaffolds designed by He et al. [267] for the bone regeneration. In another study, Makkar et al. [269] developed biocompatible and biodegradable PCL-based coatings for their potential use in orthopedic applications. Magnesium alloy (Mg) was immersed in hydrofluoric acid solution ($F_2$) which yielded $MgF_2$. **Figure 10(a)** depicts in-vitro analysis performed on the rabbit model using duplex $MgF_2$/PCL coating. The results displayed excellent cell adhesion, good cell viability, and cell proliferation, compared to uncoated Mg samples.

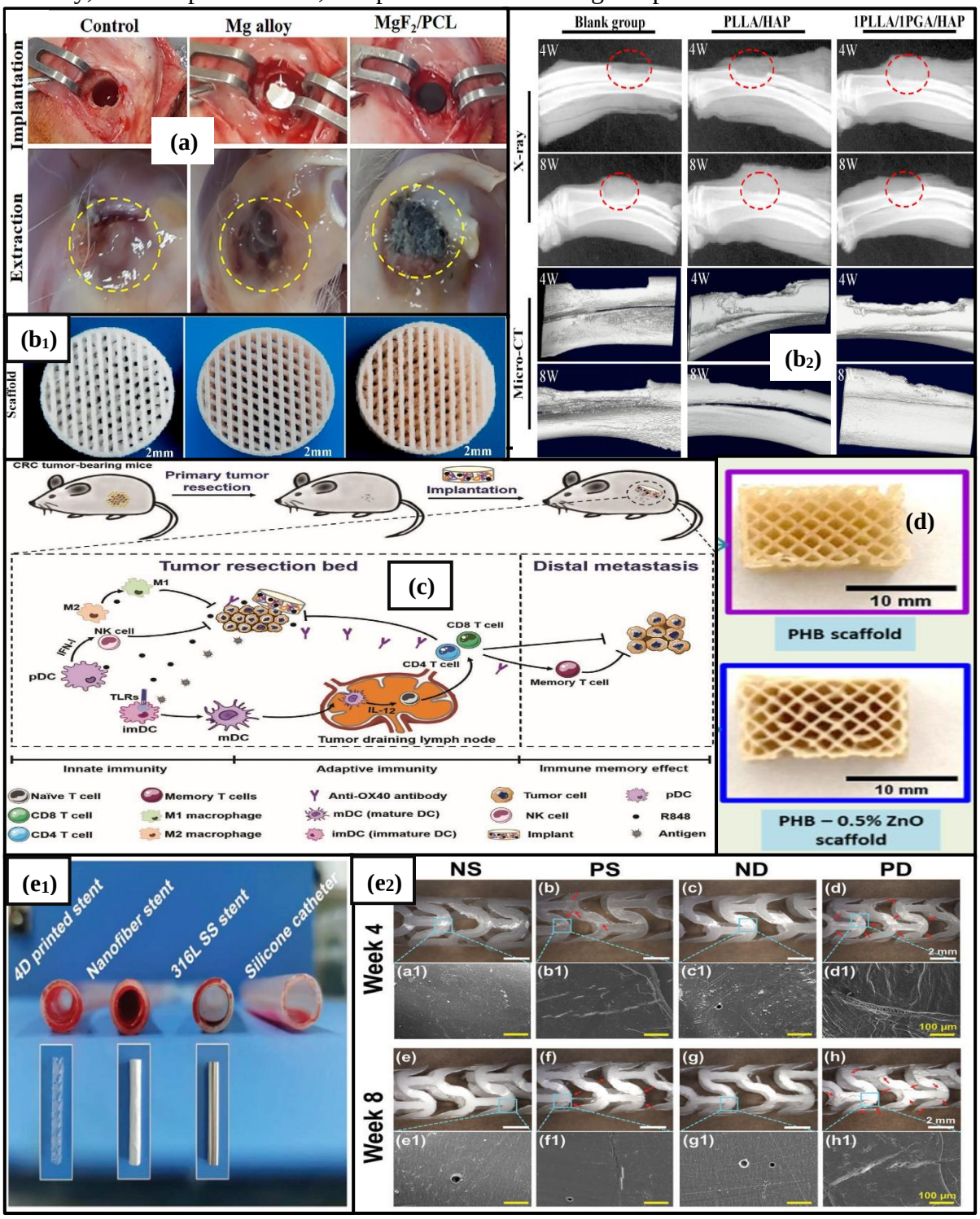



**Figure 10**. (a) Figure shows the photos of rabbit's damaged part at the time of implantation and extraction (Reproduced with permission from [269]); ($b_1$) Optical images of the PLLA (90 wt.%)/HAP (10 wt.%), PLLA (45 wt.%)/PGA (45 wt.%)/HAP (10 wt.%) and PGA (90 wt.%)/HAP (10 wt.%)

scaffolds (adapted with permission [270]); ($b_2$) X-ray radiographs and micro-CT images of the different PLLA (45 wt.%)/PGA (45 wt.%)/HAP (10 wt.%) scaffold, PLLA (90 wt.%)/HAP (10 wt.%) and blank group after four- and eight-weeks implantation. (Adapted from [270]); (c) Schematic diagram explaining the biopolymer immune implant co-loaded with R848 and anti-OX40 antibody [271] (Adapted with permission); (d). PHB- and PHB/ZnO-based composite scaffolds manufactured through 3D printing technique [272] (Adapted with permission) ($e_1$) Cross-section images of different stents models including PLA nanofiber stents after circulation for 2 hours ($e_2$) Comparison of degradation behavior of PLA stent in different environment through optical and SEM images for 4 and 8 weeks (red arrows point to the cracks) [273](adapted with permission)

Shuai et al. [270] investigated HAP/poly-l-lactic acid (PLLA)-based bone scaffolds by blending poly(glycolic acid) (PGA) through 3D printing, as depicted in **Figure 10($b_1$)** which increased its bioactivity and osteoconductivity similar to the living bone. **Figure 10($b_2$)** depicts implantation results demonstrating that the inclusion of PGA improved the degradation rate of scaffolds and weight loss increased from 3.3 % to 25 % after immersion for 28 days. Additionally, numerous pores produced by the degradation of the scaffolds helped the exposure of HAP from the matrix, which not only activated the deposition of bone-like apatite on the scaffold but also accelerated apatite growth.

Hanas et al. [274] developed electrospun PCL/HAP-coated AZ31-based composites using a friction stir process. The developed composite exhibited controlled degradation at a normal pH of 7.2 and physiological temperature. The in-vitro study showed better proliferation and adhesion as compared to uncoated scaffolds. Thus, these composites have promising nature of being used in implant applications for tissue regeneration. Likewise, Ji et al. [271] studied biopolymer-based immune implants for colorectal cancer (CRC) post-surgical therapy, as depicted in **Figure 10(c).** Immunological results demonstrated that the biopolymer immune plant treatment was performed in a two-stage action, with enhanced natural killer cell infiltration and activation of dendritic cells during the first few days. Consequently, an increased population of infiltrating T cells established immune memory effects which prevented tumor recurrence. In summary, biopolymeric sustainable composites are extensively applied for tissue regeneration of different organs due to their excellent biocompatibility, biodegradability, non-toxicity, and sustainability. Wang et al. [273] studied degradation behavior of PLA stents and their failure behavior in a dynamic condition after self-expandable deployment as depicted in **Figure 10($e_1$)**. Results showed that PLA stents exhibited excellent compression force and recovery ratio. **Figure 10($e_2$)** illustrates PLA stents complete degradation behavior in different environment and degradation conditions (NS, PS, ND and PD in **Figure 10($e_2$)** refers to no/pre deformation + static/dynamic degradation conditions respectively). PLA stents demonstrated minimal micro structural damage at 60 °C followed by 8-week degradation tests after their implantation in blood vessels. Finally, this microstructure damage triggered by deployment were responsible for acceleration in the degradation of PLA stents than fluid shear stress.

### 4.1.2. Drug Delivery Systems

Drug delivery is a core pharmaceutical research area in which biopolymers can be employed in microsphere, microcapsule, hydrogel, or implant forms. These drug delivery systems do not require any invasive procedures for removing devices after drug release [275]. Biopolymers with various drug formulations influence the drug delivery process in various ways because of several side effects and a high level of toxicity of synthetic polymers [276]. Synthetic polymers are associated with many harmful effects, including delays in treatment, and dose reduction

[277]. The controlled release of a therapeutic agent in a tunable dose is considered a prerequisite for any drug delivery system. This can be achieved through conjugation between a therapeutic agent and a drug delivery vehicle [278].

Biopolmeric materials possess extraordinary features including biocompatibility, non-toxicity, water-solubility, and biodegradability, which make them highly suitable for controlled drug delivery [279]. These drug delivery systems also prohibit enzymatic decomposition before delivering therapeutic media at the targeted locations. A variety of stimuli including chemical, physical, thermal, or combination of any of these stimuli can help the release of therapeutic medicine [280]–[282]. Biopolymeric materials in combination with HAP are often employed as potential nano-carriers. It is due to its high biocompatibility, high surface activity, high surface-to-volume ratio, and adequate biodegradability of HAP. For instance, Macha et al. [283] developed PLA/coralline HAP-based biodegradable composites for their potential use in drug delivery systems. This approach holds a good strategy to repair the defected bones using medicines and minerals through biodegradable polymer-based microcapsules. These drug delivery devices exhibited excellent ability to load and release protein, nucleic, antibiotics, and cancer drugs at a slow rate in the human body. In summary, biopolymer-based microsphere and microcapsule can encapsulate hydrophilic as well as hydrophobic drugs and regulates their targeted and controlled release at the desired location.

### 4.2 **Electronic applications**

Flexible sensing devices have been attracted worldwide due to their extensive uses in wearable sensors, smart textiles, actuators, electronic skins, flexible displays, and medical devices [284]–[286]. These devices can monitor human motions and physiological signals (referring to **Figure 11(a**)) in long term for providing clinical information for the diagnosis and prevention of diseases, as well as rehabilitation therapy. These devices are usually fabricated with synthetic polymers, metal oxides, and semiconductors [287]–[289].

Different nanofiller-based biodegradable polymer composites have found their applications, including solar-assisted cells, electromagnetic devices, and electronics [290]. The rapid growth in the daily usage of electronic devices will cause serious environmental problems due to the chemical toxicity of unused electronic devices [291]. However, the use of biodegradable polymeric composite-based devices can tackle this problem.

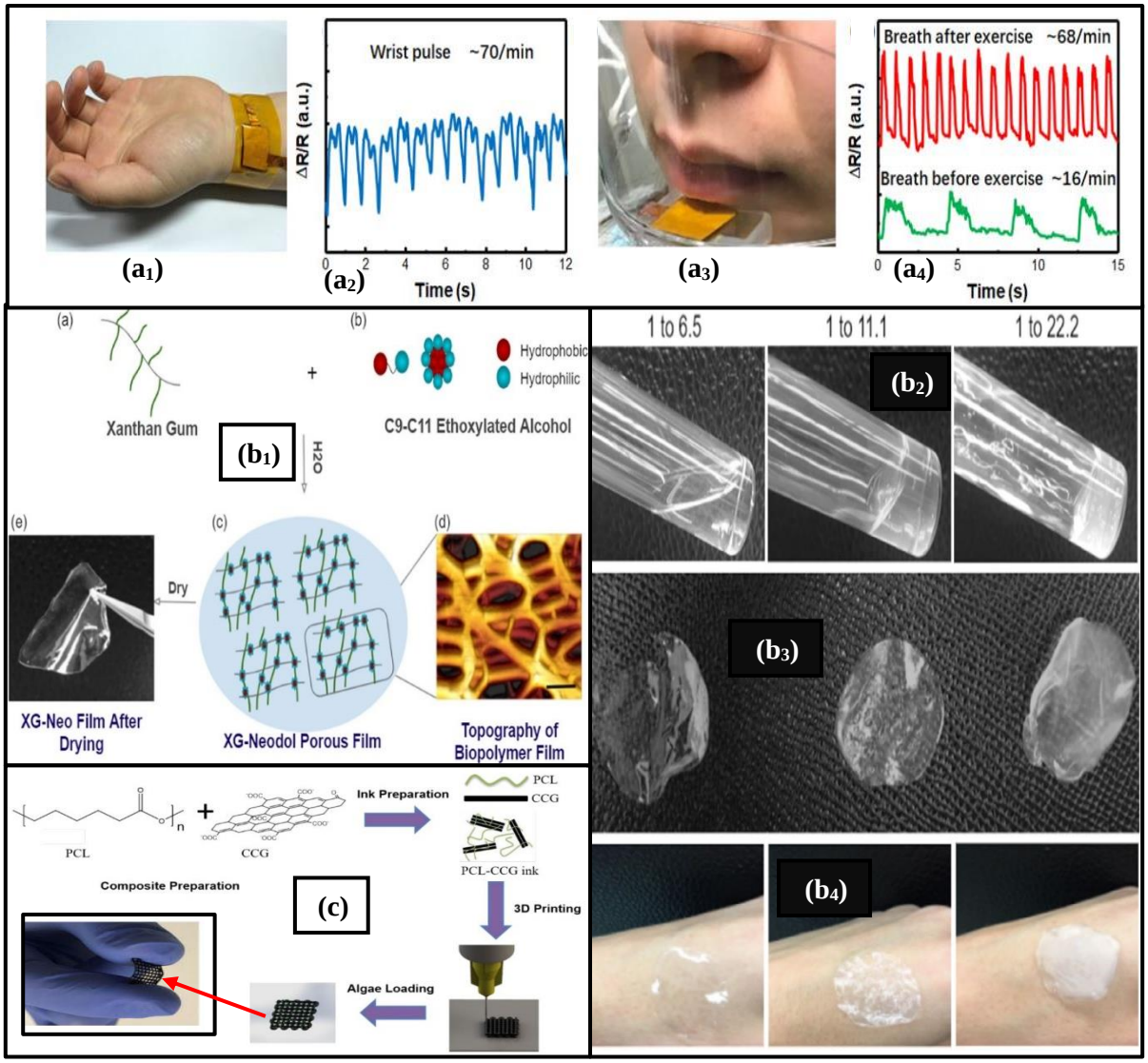



**Figure 11**. Different applications of naturally available materials in electronics; ($a_1$) Wrist pulse detection application; ($a_2$) Pulse waveform of the tester; ($a_3$) Application in respiration detection; ($a_4$) Results of breathing before and after exercise recorded by respiration detector [292] (Adapted with permission); ($b_1$) Fabrication of porous biopolymer films from the polysaccharide, xanthan gum and the ethoxylated alcohol, Neodol; ($b_2$) Macroscopic images of the viscoelastic solutions and gels before and after drying at room temperature; ($b_3$) Films fabrication after taking 500 µL of each solution and allowing to dry yields flms of different thicknesses, elasticities and transparencies; ($b_4$) Dry films placed on skin. [293] (adapted with permission); (c) Schematic diagram of procedure to form multilayer 3D-printed PCL/graphene composite electrodes, and the subsequent examination of electrochemical response via immobilization of algae (adapted with permission from [294]).

Fabijanic et al. [293] produced UV-protected film through different novel biopolymers including Neodol, xanthan gum, anionic polysaccharide, and a non-ionic surfactant, as depicted in **Figure 11(b)**. Results from electron plasmon resonance showed the free-radical reducing ability (3.5 times), while liquid chromatography-mass spectroscopy quantifies the UV-decomposition of sinapyl alcohol. Thus, this material exhibits excellent potential for its utilization as biosensors due to its inherent tenability and flexibility. In another study, PCL/chemically converted graphene (CCG)-based composite electrodes containing 10 wt.% of graphene were developed through 3D printing by Lee et al. [294]. These novel printable eco-

friendly composite electrodes are shown in **Figure 11(d)** exhibited excellent printability, processability, and robustness. Furthermore, the biocompatibility of electrodes was illustrated through electrochemical response by growing the unicellular microalgae onto PCL/CCG substrate. The printed PCL/CCG product can be potentially applied in bioelectronic applications.

### 4.3 Miscellaneous applications

The utilization of protective facemasks has increased rapidly due to the coronavirus disease 2019 (COVID-19) pandemic, and these facemasks are usually fabricated using non-biodegradable synthetic polymers, which are causing environmental problems [295]. Biopolymers are effectively applied to manufacture efficient air filters for facemasks [296]. These biopolymer-based facemasks are manufactured by incorporating nanofibers through the electrospinning process, which helps in controlling nanofiber dimensions, fiber length, and pore size [297]. Additionally, silver NPs can be used for doping fiber membranes of facemasks which permits antimicrobial characteristics as well as increases surface roughness. **Figure 12** depicts the proposed processes, biopolymers, and requirements for developing biodegradable facemasks [135].

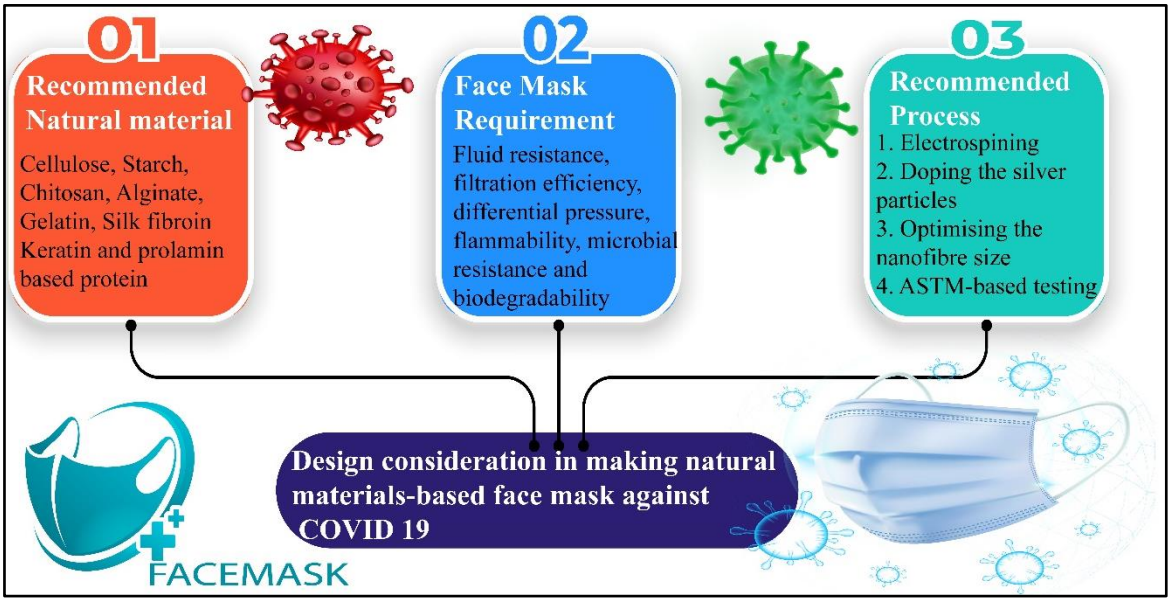



**Figure 12.** Proposed natural biopolymers, requirements, and processes for the fabrication of environmentally benign facemasks.

In the contemporary world, biopolymers are vastly applied in modern packaging and cosmetics industries due to their non-skin irritant and non-toxic nature. For instance, soy protein exhibits improved color profiles [298]. Additionally, biopolymers can be added for manicure preparation, sunscreen, scrubbing agents, and cleaning products [299]. Similarly, bivalent chelating-mineral ability and anti-microbial behavior make chitosan an excellent choice to be used as a packaging material for the preservation of edible foods [300]. Biopolymers including PLA, PHA, PBAT, and PBS exhibit comparable mechanical properties, compared to non-biodegradable polymers, and can be excellent substitutes for fabricating hygienic products and packaging materials [301].

In the textile industry, biopolymers have also gained significant attraction in the form of bed linens, towels, bandages, wipes, as well as geotextiles [302]. Superior antimicrobial properties of collagen, and alginates, permit their utilization for fabricating textile products [303].

Similarly, chitosan is commonly applied in healthcare or medical textiles for developing wound care dressings and bandages due to its biocompatibility, cationic biodegradability, and antimicrobial features. Bio-based PTT and PLA have also been employed for the manufacturing of textiles due to their extraordinary dyeability and better strength [304]. Whereas, in the agricultural industry, biopolymers mostly exist in the form of biodegradable plant pots and mulch films [305]. Similarly, the controlled release of fertilizer in the soil can be achieved using biodegradable composites [306]. **Figure 13** depicts the most recent applications of biopolymers in different engineering sectors.

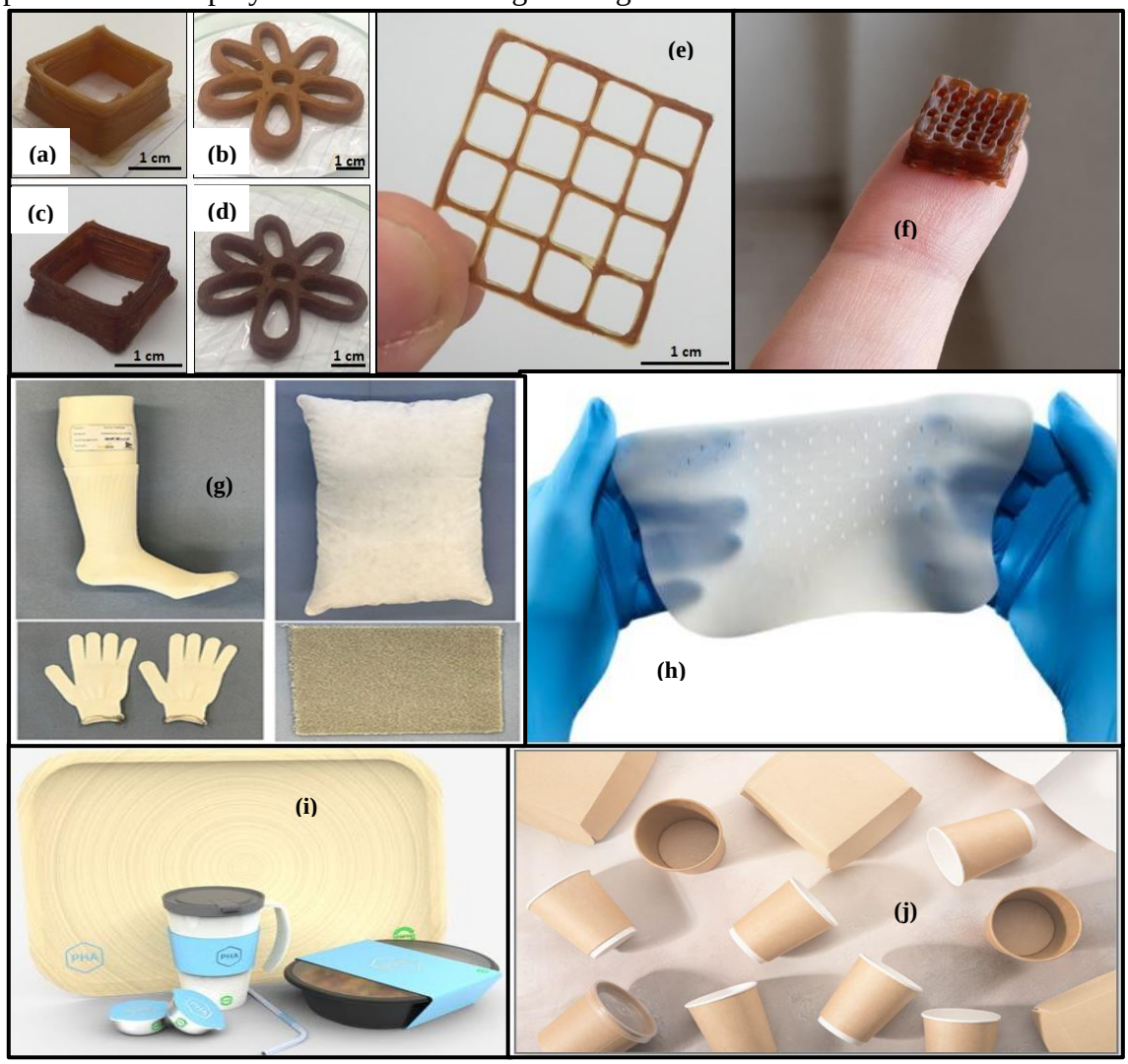



**Figure 13.** Commercially avaiable products from biopolymers; (a) Appearance of a hollow cube right after the printing process; (b) At drying conditions at ambient for 24 h; (c) A flower model before drying; (d) After drying at ambient conditions for 24 h; (e) Three-layered grid structure printed from hemicelluloses; (f) A scaffold prototype printed from hemicellulosic pastes (a-f adapted from [307]); (g) Textile products from biopolymers (@AMIBM Maastricht University 2018); (h) Collagen matrices or sponges can be used to treat wounds for tissue regrowth and reinforcement [308]; (i) PHA-based biopolymer in food packaging (according to Cambridge Consultants)

In the last few decades, there has been an increasing concern over food contamination and most fatal contaminations occur due to the presence of microorganisms [309]. Biopolymers are blended with NPs, including nanocellulose, CNTs, zinc, silver, magnesium, copper, and gold NPs, to develop biopolymeric nanocomposites [310]. Biopolymeric sustainable

nanocomposites are extensively applied in food packaging applications to extend the shelf life of food due to improved physical, thermal, mechanical, and biodegradable properties [311]. **Figure 14** depicts the various biopolymeric materials used for food packaging applications.

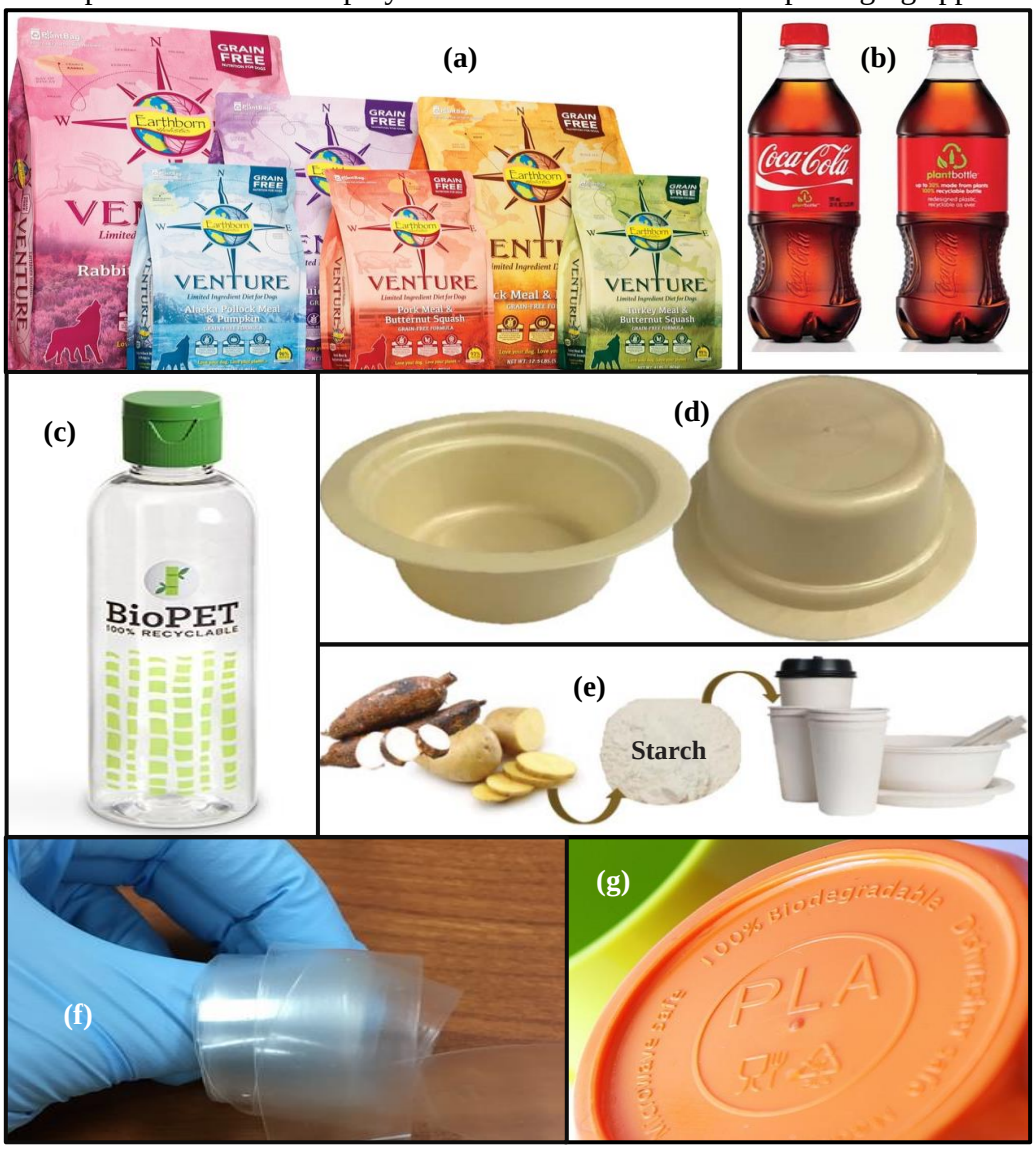



**Figure 14.** Commercially avaiable products from biopolymers; (a) The Midwestern Pet Foods-branded PlantBag containing at least 30% bio-based PE extruded with fossil fuel-based PE (adopted from [312]); (b) Image of the PlantBottleTM made up to 30% from biomass and 100% recyclable. (Picuture Courtesy of the Coca-Cola Company (Atlanta, USA); (c) 100% Recyclable Bio-PET Bottle (adopted from [313] ); (d) Biodegradable food tray made of PHB obtained by injection molding (adopted from [314]); (e) Biodegradable packaging articles based on starch [314]; (f) CNC-based biopolymers for packaging applications (adopted from [315]); (g) PLA based products for pakaging (adopted from [316])

## 5. Future challenges and opportunities

Despite the tremendous progress made in the field of biodegradable biopolymers, some areas of research still require further exploration for addressing challenges. Biopolymers are intriguing raw materials for promoting eco-friendliness, non-toxicity, and sustainability. There is a need to conduct focused research for improving the mechanical characteristics and biodegradation mechanisms of complex working environments. Additionally, biopolymers are beneficial for the ecological environment only if their degradation products are non-toxic. To

date, no study has been found in the literature that evaluates the harmful and toxic impact upon the biodegradation of biopolymers.

Furthermore, several weaknesses in some biopolymers including poor processability and solubility in water or common organic solvents have been observed. These areas need to be explored to encourage the utilization of biopolymer materials. Biodegradable packaging for agricultural products also needs more attention in terms of gas barrier properties and water resistance. In biomedical areas, tissue biomimicry still remains a key challenge. Thus, more development and focus on 3D printing of biopolymers-based tissues or organs are required. This is because of some challenges related to material printability, the exact geometry of organs, functionality, and safety (biosafety and environmental safety). **Figure 15** summarizes the challenges in terms of application requirements that need attention in the future.

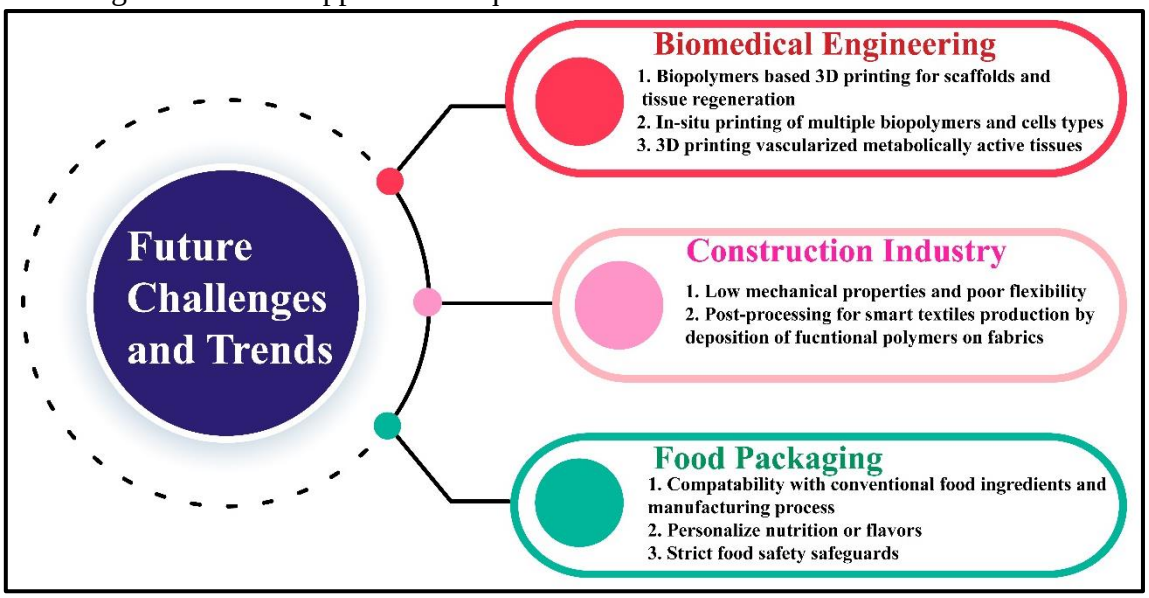



**Figure 15**. Future challenges and trends for biopolymers.

## 6. Conclusions

Recent advancements toward promoting a sustainable environment require the utilization of green materials, especially biopolymers and their composites, which possess the ability to replace synthetic polymers due to their extraordinary biodegradability, processability, renewability, biocompatibility, and non-toxicity. Besides these advantages, these biopolymers also have excellent technological and economic perspectives. The developed biopolymeric composites were characterized through spectroscopic, microscopic, diffraction, and degradation testing. These biopolymeric composites have exhibited their efficiency and integrity to be applied in textiles, implanting devices, tissue engineering, drug delivery systems, and agricultural industries. Non-biodegradable polymer composites are usually preferred over biodegradable biopolymer composites due to their superior mechanical characteristics. However, these plastics cannot be degraded and are severely influencing the ecological environment. Different biopolymers, including PLA, PHA, cellulose, and other naturally occurring biopolymers and their composites, demonstrated excellent degradation behavior, implying that these advantageous biopolymers will not only help to reduce the accumulation of plastic waste, but will also help to reduce the global warming effect.

The world needs to shift its attention toward the environmental pollution problem caused by plastic waste and its possible remedies. Reasonable and comparable mechanical characteristics of biodegradable biopolymer composites help their possible utilization in all engineering fields

to produce a cleaner and greener environment by cutting down on the accumulation of plastic waste.

## Conflict of interest statement

The authors declare no conflict of interest.


## Funding

This work was not supported by any funding.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